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Lead release to potable water during the Flint, Michigan water crisis as revealed by routine biosolids monitoring data

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A B S T R A C T

Routine biosolids monitoring data provides an independent and comprehensive means to estimate water lead release pre-, during and post-Flint Water Crisis (FWC). The mass of potable plumbing-related metals (i.e., lead, cadmium, copper, nickel and zinc) in sewage biosolids strongly correlated with one another during the FWC ($p < 0.05$). A simple parametric regression model based on 90th percentile potable water lead measurements (WLL90) from five city-wide citizen science sampling efforts August 2015–August 2017 was strongly correlated to corresponding monthly lead mass in biosolids [Biosolids-Pb (kg) = 0.483 x WLL90 (μg/L) + 1.79; $R^2 = 0.86$, $p < 0.05$]. Although total biosolids lead increased just 14% during the 18 months of the FWC versus the comparable time pre-FWC, 76% of that increase occurred in July–September 2014, and the corresponding percentage of Flint children under 6 years with elevated blood lead ≥ 5 μg/dL (i.e., %EBL5) doubling from 3.45% to 6.61% in those same three months versus 2013 ($p < 0.05$). %EBL5 was not statistically higher during the remaining months of the FWC compared to pre-FWC or post-FWC. As expected, lead in biosolids during the FWC, when orthophosphate was not added, was moderately correlated with water temperature ($R^2 = 0.30$, $p < 0.05$), but not at other times pre- and post-FWC when orthophosphate was present. Tripling the orthophosphate dose post-FWC versus pre-FWC and some lead pipe removal, decreased lead in biosolids (and %EBL5) to historic lows (2016–2017 vs. 2012–2013; $p < 0.05$), supporting the effectiveness of these public health interventions in reducing childhood water lead exposure.

1. Introduction

After a citizen science collaboration between Flint residents and our Virginia Tech research team exposed citywide water lead contamination in August 2015, there has been widespread concern regarding consumer exposure to lead during the Flint Water Crisis (FWC) (Bellinger, 2016; Edwards, 2015; Pieper et al., 2017, 2019; Roy and Edwards, 2019a, 2019b). The FWC started April 25, 2014, when Lake Huron water from Detroit with a 1 mg/L orthophosphate dose for corrosion control was replaced with higher corrosivity Flint River water without orthophosphate. This change increased lead release from lead service lines (LSLs), galvanized iron pipe (GIP), leaded solder and brass (Del Toral, 2015; Edwards et al., 2018; Masten et al., 2016; Pieper et al., 2017, 2018).

Although the trajectory of water lead levels (WLLs) in the home of one Flint resident is relatively well understood (Pieper et al., 2017), almost nothing is known about the magnitude and timing of lead release throughout the rest of the city between April 2014 and August 2015 – the official water lead data that was collected is considered nearly useless due to the fact compliance samples did not meet criteria under the National Primary Drinking Water Regulations and the federal Lead and Copper Rule (LCR). For example, the requirement that at least 50% of compliance samples must be collected from homes with LSLs was not met (Pieper et al., 2018). The official sampling results were also biased low, by pre-flushing homes the evening before the six-hour stagnation time and use of small-mouthed bottles (Edwards, 2015; Grevatt, 2016; Milman and Glenza, 2016; Del Toral, 2015). In contrast, starting with Virginia Tech's standardized citywide sampling campaign in hundreds of Flint homes August 2015 onwards, there is relatively good understanding of WLL trends because these sampling events were repeated at the same homes using the same sampling protocol in March 2016, July 2016, November 2016 and August 2017 (Pieper et al., 2018).

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The lack of data on water lead levels and associated uncertainties with human exposure from April 2014–August 2015 have led to angst, speculation, proxy research and controversy (Bouffard, 2018; Carmody, 2019; Clark and Filardo, 2018; Gómez and Dietrich, 2018; Haynes, 2019; Taylor et al., 2016; Roy, 2017). The use of routine childhood blood lead level (BLL) monitoring data to indirectly assess the severity of the water lead exposure (e.g., Hanna-Attisha et al., 2016; Kennedy, 2016; Zahran et al., 2017; Gómez et al., 2018; Gómez et al., 2019), relies on a dataset of children mostly 1.5–6 years age, who are actually the group least likely to reveal water lead impacts because this group is at greatest risk of exposure to lead paint and dust (Levin et al., 2008; Edwards et al., 2009; Triantafyllidou and Edwards, 2012; Hanna-Attisha et al., 2016). The pregnant women and formula-fed infants who are at greatest risk of lead exposure from drinking water (i.e., >85% of lead exposure for many infants fed reconstituted formula is typically from water) do not have routine blood lead measurements for analysis because it is assumed that federal corrosion control laws would limit water lead exposures (Edwards et al., 2009; Edwards, 2013; Triantafyllidou and Edwards, 2012; USEPA, 1988; USEPA, 1991).

Despite the acknowledged weaknesses in the elevated blood lead (EBL) data used in prior ecological assessments of the FWC and some heated debate about their interpretation, there is good agreement about the overall trends (Gómez et al., 2018; Hanna-Attisha, 2018a,b; Kennedy, 2016; Zahran et al., 2017). Specifically, the proportion of Flint children with elevated blood lead levels ≥ 5 µg/dL (%EBL5) roughly doubled during the FWC (April 2014–October 2015), especially in the neighborhoods where Virginia Tech's water sampling revealed greatest lead in water risk (Gómez et al., 2018; Hanna-Attisha et al., 2016). However, there is nonetheless ongoing dismay about "not knowing" trends in water lead exposure that occurred during the 18 months of the FWC (Banner, 2018; Gómez et al., 2018; Gómez et al., 2019; Graham, 2016; Kruger et al., 2017; Oleske et al., 2016).

To address these concerns, we explored the novel hypothesis that routine monthly analysis of metal mass in biosolids (i.e., digested sewage sludge) at the Flint wastewater treatment plant represents a composite sample tracking the mass of metal release from plumbing to the Flint water distribution system. Metals (including lead) in municipal wastewater are often dominated by release from potable water plumbing (Alam and Sadiq, 1989; Boulay and Edwards, 2000; Comber and Gunn, 1996; Isaac and Boothroyd, 1996; Hargreaves et al., 2018; Karvelas et al., 2003; Murphy and Pierides, 2004; NYCDEP, n.d.; Santos-Echeandía, 2009; Sörme and Lagerkvist, 2002; Toffey, 2016; Goodman, 1984; Isaac et al., 1997) and a majority of Pb ($87 \pm 8\%$) in wastewater is typically removed during treatment and concentrated in the biosolids (Goldstone et al., 1990; Goldstone and Lester, 1991). This general idea was recently exploited in attempts to estimate water lead levels in drinking water of ancient Rome (Delile et al., 2014, 2017). Biosolids in wastewater began to be monitored in the U.S. starting in early 1980s and, therefore, this dataset is available for many municipal systems starting before the Lead and Copper Rule monitoring began in 1991 (USEPA, 1991; WEF, 2011). In Flint less than 5% of the wastewater is derived from industry, which has largely eliminated its lead sources (Case, 2018), further increasing the likelihood that the lead captured in Flint biosolids is mostly derived from domestic plumbing release to potable water.

If the hypothesis is valid that lead release from potable water plumbing is a substantial source of lead in Flint biosolids, it would also be expected that:

a) metal mass released from plumbing and captured in biosolids would be higher during the FWC (especially in warmer months) when orthophosphate corrosion inhibitor was absent, but lower and independent of temperature during time periods when orthophosphate was present (Boulay and Edwards, 2000; Del Toral et al., 2013; Deshommes et al., 2013; Masters et al., 2016b; Lytle and Schock, 1996).
b) Lead in biosolids will correlate with other common metals characteristic of premise plumbing materials (cadmium, copper, zinc and nickel) that sloughed from pipe scale when corrosion control was interrupted (Alam and Sadiq, 1989; AWWA, 2011; Comber and Gunn, 1996; Gonzalez et al., 2013; Lytle and Schock, 1996; Pieper et al., 2017).
c) lead mass in biosolids would correlate with available datasets collected for citywide lead in water, and possibly the incidence of %EBL5 in Flint children during the anomalous time when lead in water was a dominant source of childhood lead exposure (Table S1).

2. Experimental methods

Water temperature data, %EBL5 cases for children under six years of age within the City of Flint, MI, and pre-existing data on metal concentrations in biosolids were used in this retrospective ecological study analyzed for three periods (Table S2): May 2011–April 2014 ("pre-FWC"), May 2014–October 2015 ("during FWC") and November 2015–November 2017 ("post-FWC"). The term "post-FWC" for lead, reflects the fact that bottled water and filters were provided to all residents for health protection after November 2015, reducing the likelihood of consumer exposure even as water lead remained elevated. This also coincided with a switchback to Lake Huron water and boosted orthophosphate dosing that began to reduce water and biosolids lead.

2.1. Water temperature

Daily temperature data at the effluent of the Flint water treatment plant was obtained from archived monthly Michigan Department of Environmental Quality water quality reports 2011–2017 (https://www.michigan.gov/flintwater/).

2.2. Trends in elevated blood lead (EBL)

Deidentified summaries of BLL measurements for Flint children were provided by Hurley Medical Center's Dr. Mona Hanna-Attisha for May 2011–November 2017, and used to calculate percentage of children under six with elevated blood lead levels (%EBL5) using conventions described elsewhere (Hanna-Attisha et al., 2016; Hanna-Attisha, 2018a,b). To account as best we can, for a slight lag between increased WLLs and elevations of lead in children's blood considering the blood lead half-life of 28–36 days (ATSDR, 2007; Triantafyllidou and Edwards, 2012), correlations used %EBL5 data paired with biosolids lead from the same month (i.e., %EBL5 cases detected May 2016 are paired with biosolids data from May 2016).

2.3. Metals in biosolids

Monthly metal concentrations in biosolids (lead, cadmium, copper, nickel and zinc; mg/kg on a dried weight basis) and total monthly biosolids production (kg) were provided by MDEQ. A composite sample of biosolids was collected from an effluent digester pipe at the City of Flint wastewater plant (Case, 2018) early each month from May 2011 to November 2017. Seventy seven percent of the biosolids samples were collected between the 1st and 6th of each month. The metal concentrations were measured per Standard Method SW 6020A (APHA, AWWA and WEF, 1998).

The monthly mass of metal in biosolids was estimated by multiplying the metal biosolids concentration by the total biosolids production.

A one month offset was used between WLL90 data from Virginia Tech's citywide sampling campaigns with metals in biosolids data, to partly account for the two weeks (plant target = 13–15 days) of biosolids retention time (WEF, 2011; Case, 2018) in the plant digester and another few days of activated sludge detention time. For example, water samples collected from homes throughout the month of August 2015 were paired with total lead mass in biosolids early September 2015.

2.4. Metals in water, including water lead levels (WLLs)

Five city-wide tap water sampling events using a three-bottle first, second and third draw protocol were executed in a citizen science collaboration between Flint residents and our Virginia Tech team in August 2015 (n = 268), March 2016 (n = 186), July 2016 (n = 176), November 2016 (n = 164) and August 2017 (n = 150) and the results were published elsewhere (Pieper et al., 2018). All water samples were acidified by adding 2% HNO_3, digested for 16 + hours to adequately dissolve and capture particulate lead and analyzed on an Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for 28 elements including lead, cadmium, copper, nickel and zinc (all μg/L) per Standard Method 3125 B (APHA, AWWA and WEF, 1998). For data quality assurance and quality control, blanks and spikes of known concentrations were measured on the ICP-MS after every 10–15 samples.

Sequential samples of lead, copper and zinc in water of 23 Flint homes that participated in all five USEPA sequential sampling rounds (January–March, May, July, September, and November 2016) during the 2016 federal emergency response were obtained from the USEPA's website (USEPA, 2017; Lytle et al. 2019).

Mean composite metal sample values using the weighting [1/3 x (mean first draw) + 1/3 x (mean second draw) + 1/3 (mean third draw)] were calculated in the five Virginia Tech and the five USEPA sampling rounds to test co-occurrence of zinc, lead and copper. Similar calculations could not be made for nickel and cadmium in the Virginia Tech data because the mean release was below the detection limit.

A representative weighted average 90th percentile WLL (WLL90) was calculated, using a weighted average of 1/3 x (90th percentile first draw lead), 1/3 x (90th percentile second draw lead) and 1/3 x (90th percentile third draw lead), to reflect importance of lead release from all three types of water in human exposure (Sandvig et al., 2008) for each Virginia Tech sampling round. The 90th percentile has been the standardized reporting measure of WLL in the United States since the federal Lead and Copper Rule was adopted in 1991.

For a comparison between the WLLs in Washington D.C 1997–2006 and the FWC, a modified composite 90th percentile WLL (MWLL90) was calculated based on reported first draw lead levels from our previous studies, because no third draw data and only one set of extensive second draw WLLs exists for Washington D.C (Edwards et al., 2009; Edwards, 2013). Specifically, to characterize human exposure risk to water lead, we used a 50:50 weighting of measured 90th percentile first draw and measured 90th percentile second draw for the FWC, using a viable LCR sampling pool comprised of 50% homes with LSLs (17 homes with LSLs and 17 homes with lead solder/galvanized iron) back-calculated as described in Pieper et al. (2018). For Washington D.C., we estimated the corresponding weighted 90th percentile water lead level (50:50 first and second draw weighting), using our previously published 90th percentile first draw lead calculations, and an assumption that the 90th percentile second draw lead was equal to 1.375 × 90th percentile first draw lead, per calculations of 6,162 first and second draw samples collected during summer 2003.

2.5. Statistical analyses

All statistical analyses were conducted in RStudio (version 3.3.2) and/or Microsoft® Excel® (version 2016). A p value of <0.05 with an alpha value (α) of 0.05 was selected to determine statistical significance. The Pearson's coefficient correlation test was used to examine the associations between monthly biosolids lead mass, % EBL5 and other variables. Parametric linear regressions were performed between: a) composite WLL90 from all five Virginia Tech sampling rounds (μg/L) and lead in biosolids (kg) in the corresponding month, b) MWLL90 from all five Virginia Tech sampling rounds (μg/L) and lead in biosolids (kg) in the corresponding month, and c) %EBL5 for 18 months of the FWC and lead in biosolids (kg) in the corresponding month. The runs test for randomness was conducted for lead in biosolids (kg) for 18 months of the FWC (NIST/SEMATECH, 2012).

3. Results and discussion

3.1. Temporal trends of plumbing-related metals in biosolids

Levels of premise plumbing related metals (cadmium, copper, lead, nickel and zinc) in Flint's monthly biosolids were correlated before, during and post-FWC (Figs. 1 and S1). All five metals spiked markedly in the summer of 2014 when there was no corrosion control during the FWC, and it is hypothetically possible this was due to general sloughing of pre-existing scale from all plumbing surfaces (Masten et al., 2016; Olson et al., 2017; Pieper et al., 2017, 2018). The mass of all five plumbing-related metals in biosolids were strongly correlated ($p < 0.05$) with each other in all three time periods (Table S3). At the home of Flint "Resident Zero", there was a similar correlation between all five of these plumbing related metals during intensive water sampling in April 2015 (Pieper et al., 2017), when this home pipe scale was clearly sloughing at high levels—hence, the system wide correlation is a logical extension of what occurred in this one home.

Considering the five Virginia Tech citywide potable water sampling rounds, strong correlations were observed between the mean composite measures of lead and copper ($R^2 = 0.89$, $p < 0.05$), and copper and zinc ($R^2 = 0.83$, $p < 0.05$) (Table S4). Similar trends were observed for all five rounds of USEPA data, with lead and copper ($R^2 = 0.93$, $p < 0.05$), and copper and zinc ($R^2 = 0.78$, $p < 0.05$). Interestingly, there was no significant co-occurrence observed for these metals for a given draw in both datasets (i.e., first, second or third draw), but only for the weighted composite result. This is expected given that the first draw water is often derived from a pure copper pipe, whereas the second draw sample is often from a service line with pure lead or galvanized iron pipe (i.e., first draw has highest copper and relatively low lead, second draw has highest lead and almost no copper).

The monthly lead in biosolids was not correlated with monthly average water temperature pre- and post-FWC ($p > 0.05$) as expected for systems dosed with orthophosphate for corrosion control (Masters et al., 2016b), but they were moderately correlated during the FWC when orthophosphate was not dosed ($R^2 = 0.30$, $p < 0.05$) (Table S1). The water temperature of the shallow Flint River water source versus Lake Huron, fluctuated much more in both summer and winter months during FWC compared to pre- or post-FWC (Fig. 1), producing a greater possible effect of temperature on metal release from plumbing (Deshommes et al., 2013; Masters et al., 2016b).

The tripling of orthophosphate dose to Lake Huron water (3 mg/L

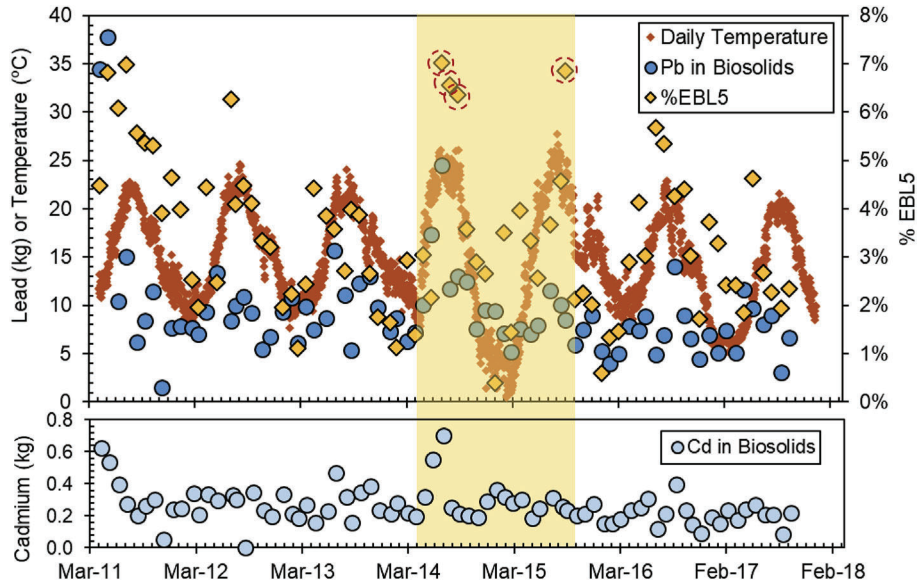

Fig. 1. Monthly cumulative lead (Pb) mass in biosolids, percent of children with elevated blood lead levels (% EBL5) (i.e., ≥5 μg/dL) and water temperature (upper) during May 2011–November 2017. The four months with peak %EBL5 during the FWC (circled in red) occurred in July–September 2014 and August 2015. Representative data for a plumbing related metal (Cd), also illustrates prominent spike immediately after the water switch to Flint River in April 2014 (lower). The light orange area represents the time span of the FWC. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

total PO_4) starting December 9, 2015 per USEPA recommendations decreased overall lead release from the distribution system to historic lows post-FWC (2016–17; average lead in biosolids = 6.9 kg/month) versus pre-FWC (2012–13; average = 9.3 kg/month), suggesting that the higher phosphate dosages caused lower lead release from the plumbing even after the interrupted corrosion control during the FWC (two-tailed paired *t*-test; $p < 0.05$). This trend of declining potable lead levels was confirmed by third party independent sampling which revealed a 90th percentile first draw WLL of 4 μg/L in late January 2019 (Masten and Doudrick, 2019).

3.2. Biosolids lead correlates strongly with citywide water lead measurements

The WLL90 calculated from each round of citywide citizen science sampling and the monthly lead in biosolids of the corresponding month were strongly correlated ($R^2 = 0.86$, $p < 0.05$, N = 5; Fig. 2), supporting the hypothesis that biosolids lead reflects citywide release of lead to water from plumbing. The modeled relationship [Biosolids-Pb (kg) = 0.483 x WLL90 (μg/L) + 1.79], has an intercept of 1.79 kg, which might represent a portion of lead loading to the sewage plant per month independent of that

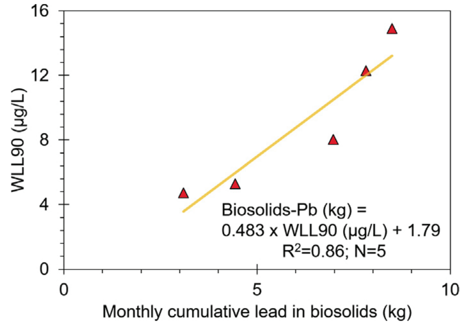

Fig. 2. Monthly cumulative lead mass (kg) in biosolids was correlated with WLL90 from five rounds of water sampling campaigns ($R^2 = 0.86$, $p < 0.05$, N = 5).

released from plumbing, such as lead in wastewater from all non-plumbing sources and 5% wastewater flow from industry (Case, 2018).

Recent research made a rough estimate that ~18 g lead leached on average from each Flint LSL (Olson et al., 2017). Considering a recent rough estimate of 12,000 homes with LSLs in the City of Flint (City of Flint, 2019), a total extra lead release of 216 kg from LSLs to water would be predicted during the FWC. If the typical 87% of this lead release to water was captured in biosolids (Goldstone et al., 1990; Goldstone and Lester, 1991), the resulting prediction of 188 kg lead is of similar magnitude to the 184 kg cumulative lead measured in biosolids during the FWC in this research.

In terms of possible confounding factors, the stormwater in Flint is not discharged to sewers, reducing the likelihood that surface water runoff or hydrant flushing of water would influence the results (Busch, 2014; Emery, 2014; Fonger, 2014; Roy and Edwards, 2015; Case, 2018). Moreover, hydrant flushing uses water from mains that has not contacted the building plumbing that contains the Zn, Cu, Pb, Cd and Ni metals, so any variation in flushing water from hydrants does not affect the release of these metals to drinking water or their mass in biosolids. Following the switch, the total mass of biosolids (dry weight basis) produced in May to July 2014 (average = 317 metric tons) was more than twice as high as the biosolids produced the prior year (May 2013–Apr, 2014; average = 140 metric tons), before eventually stabilizing to pre-crisis levels after switching back to Lake Huron source water (Fig. S2). As long as the unidentified source of these higher biosolids did not contain a significant mass of lead, the correlation between lead mass in biosolids and the lead release to potable water would still be valid, as appears to be the case for data presented herein.

3.3. Lead in biosolids and elevated blood lead

Lead in water exposure is not typically considered to be a dominant correlate to lead in blood (Triantafyllidou and Edwards, 2012), especially for the age group whose blood lead is routinely monitored, when corrosion control is effective or the population is protected against elevated lead in water. This expectation was

verified by a lack of correlation between biosolids lead (reflecting water lead exposure) and %EBL5 post-crisis ($p > 0.05$; Table S1). During the 18 months pre- and during the FWC, there were only very weak correlations between %EBL5 and biosolids lead (during FWC, $R^2 = 0.22$; pre-FWC, $R^2 = 0.12$; $p < 0.05$).

Overall, it appears that monthly lead mass in biosolids does seem to track lead release from plumbing to potable water, but biosolids lead is only slightly predictive of overall childhood exposure for the age group of children whose blood lead is tested and considered least sensitive to water exposure. Several decades ago, it was predicted that the typical 2-year-old child obtained only 20% of their lead exposure from water on average, with the remainder from food, dust and other sources (USEPA, 1988). The fact that there are only slightly significant correlations between biosolids lead and %EBL5 for this age group both pre-FWC and during the FWC, suggests that lead in water is not normally a dominant source of exposure for this age group, even though other sources of lead exposure have been increasingly controlled since 1990 when the 20% blood lead from water estimate was made (Triantafyllidou and Edwards, 2012; USEPA, 1988).

The overall downward trends of lead in biosolids and childhood lead exposure are generally consistent with those observed nationally (Gómez et al., 2018; Hargreaves et al., 2018; Toffey, 2016; USEPA, 2009). Lead in biosolids was in a clear downtrend during the pre-FWC time period from 12 kg/month (May 2011–October 2012) to 9 kg/month (November 2012–April 2014). This downtrend is also reflected in decreasing mean %EBL5 from 4.71% to 2.76% in Flint over the same time period similar to trends nationwide (CDC, 2018; Gómez et al., 2018).

There are only two major exceptions to the overall %EBL5 and biosolids lead downtrend. The first was pre-FWC in 2011, where an unexplained peak in monthly biosolids lead correlated to a peak in %EBL5. Specifically, the average lead in biosolids for May–October 2011 of 18.7 kg/month corresponded to a very high mean %EBL5 of 5.91% over that time period. Gomez and colleagues (2018) attributed the 2011 spike in %EBL5 to a "random variation," but the biosolids data indicate that lead in wastewater was also anomalously high in this April-May 2011 time period well before the FWC began in 2014. Looking more closely at the 2011 biosolids lead spike, we note that the lead levels were 115–185% above the Ni trendline and 30–70% above the Cd trendline, and during this time period lead was not correlated with other metals originating from premise plumbing corrosion (Fig. S3a). We speculate that this anomaly may have somehow been linked to treatment upsets or other events during record Detroit rainfall, which was national news in that exact time period (Bienkowski, 2013). ***Regardless, a key point is that both biosolids lead and %EBL5 spiked higher in 2011 than at any other point reported in this research, including during the FWC.***

The second exception occurred shortly after the source water was switched to Flint River in April 2014. The total biosolids lead mass during 18-month intervals before, during FWC and after the FWC were 161.5 kg, 184 kg, and 129 kg, respectively. Of the total 23 kg (or ~14% overall increased mass) extra biosolids lead mass during versus pre-FWC, 76% came in July–September of 2014 versus July–September 2013 (Fig. 3). A runs test for randomness analysis confirms that this biosolids lead mass spike in 2014 was not random ($p < 0.05$). The corresponding %EBL5 roughly doubled from 3.45% to 6.61% in those three months of 2014 versus 2013 ($p < 0.05$), and this was also the only time period in which %EBL5 was statistically higher during the FWC than pre- or post-FWC (Fig. 3). This 2014 biosolids lead spike was also directly on the trendline for other biosolids metals derived from plumbing materials (e.g., Ni and Cu - Fig. S3b), reinforcing the belief that this spike in biosolids lead (and associated spike in %EBL5) was due to scale

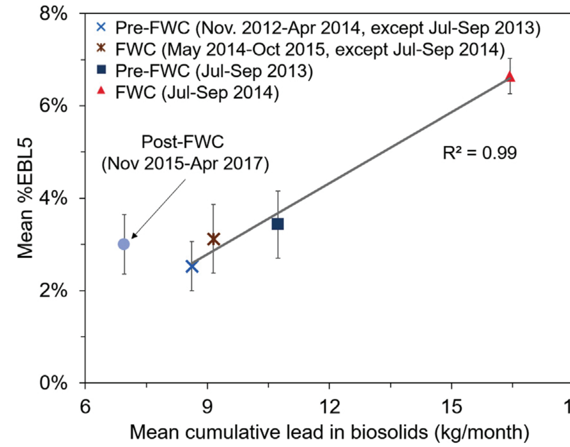

Fig. 3. Mean cumulative lead mass in biosolids (kg/month) correlated with mean % EBL5 for four time intervals pre- and during FWC ($R^2 = 0.99$, $p < 0.05$). Error bars indicate 95% confidence intervals for %EBL5. Due to water protective measures and a dramatic increase in EBL testing frequency by Federal Emergency Management Agency (FEMA), the post-FWC result is excluded from the regression.

sloughing from plumbing. In the 18 months post-FWC, the total lead in biosolids dropped 30% (~55 kg) and mean %EBL5 was 3.00% (Fig. 3).

After Virginia Tech's drinking water advisory in August 2015 and then Hurley Medical Center's September 2015 press conference showed increased blood lead in Flint children, the water source was switched back to Lake Huron water with corrosion control and decisive public health interventions were implemented to protect the public from high WLLs (Hanna-Attisha et al., 2016; State of Michigan, 2016; Pieper et al., 2018). The lead in biosolids decreased to 7.5 kg/month and mean %EBL5 decreased to 3.12% over the following year (November 2015–October 2016).

With recovery of corrosion control in the distribution system and implementation of enhanced corrosion inhibitor dosing (~3 mg/L as PO_4 starting December 9 2015), the lead in biosolids and mean %EBL5 was further decreased to historical lows of 6.7 kg/month and 2.58% (November 2016–October 2017), respectively. Clearly, the public health interventions of bottled water and lead filters reduced %EBL5 incidence back to historical lows, and %EBL5 and biosolids lead were decoupled, even as WLLs remained above federal standards through June 2016 as indicated by both the State of Michigan official data on residential/sentinel sampling and Virginia Tech's citizen science water lead monitoring (MDEQ, 2018; Pieper et al., 2018).

3.4. Contrast and comparison to other data

If it is assumed that the net biosolids lead minus the baseline 1.79 kg of non-plumbing lead (Figs. 2 and S4) reflects the true trend in water lead release and exposure, a perspective is provided on the FWC that is remarkably consistent with most existing published research on WLL or using the %EBL5 proxies. After the switchback in October 2015, the official lead in water data started meeting federal standards in late 2016· and the corresponding lead in biosolids also declined back to levels considered normal at the start of this decade (Fig. S5). Blood lead levels and water lead levels have also recently dropped to historic lows in Flint (Gómez et al., 2018; MDEQ, 2018; Pieper et al., 2018; Gómez et al., 2019; Masten and Doudrick, 2019; Lytle et al., 2019).

These results also help address some speculation regarding changes in childhood lead exposure in Flint. Specifically, the 2014 blood lead spike in Flint children did not likely originate from

children having more contact with contaminated soil in summer months as was recently speculated (Laidlaw et al., 2016), because the high lead in biosolids co-occurred with release of lead from premise plumbing. Thus, our data supports recent analyses by Centers of Disease Control and Prevention (CDC) and others, that also indicated soil was not a major contributor to this 2014 blood lead spike based on independent reasoning (Kennedy, 2016; Sadler et al., 2017). Moreover, speculation that a drop in blood lead observed in the months of Apr-Sep 2014 to Sep 2014–Sep 2015 is due to “boil water advisories” that caused consumers to switch to bottled water (Zahran et al., 2017), is not necessary because the drop in %EBL5 is shown herein to reflect a drop in WLLs in that time period.

Finally, this analysis provides evidence that the public health interventions of lead filters and bottled water were highly effective, severing any link between %EBL5 to lead in water and biosolids ($p > 0.05$, Table S1).

3.5. Historical perspectives on the Flint Water Crisis

This analysis fills major knowledge gaps regarding the trajectory of the FWC in relation to lead in water and human exposure. In particular, the monthly lead in biosolids reached a peak of 24.5 kg during the warmer months (May–October) of the crisis in 2014, but lead release steadily declined thereafter to less than half of that value (11.5 kg) for the same time period in 2015. Moreover, the average and maximum biosolids lead measurements during the FWC in 2015 were comparable to those pre-FWC in summer 2012 and summer 2013, suggesting that WLLs throughout the city might have declined from the start of the FWC in summer 2014 as lead was depleted and sloughed from scale (Olson et al., 2017). This analysis strongly suggests that the “worst” lead exposure during the FWC was restricted to June–August 2014 (captured in biosolids lead mass during July–September 2014), as is further confirmed by the significant elevation in %EBL5 associated with those months. The overall biosolids lead data directly contradicts prior speculation by ourselves and others, that water lead levels and associated exposures, progressively increased during the 18 months of the FWC.

Our analysis can also help put the potential exposures occurring during the FWC into context, versus routine USEPA 90th percentile first draw lead levels that are reported in other cities, and also in comparison to the other major water lead contamination event of this century in Washington D.C. 2001–2004 (Roy and Edwards, 2019a). Virginia Tech's citywide sampling event in August 2015, did detect a very significant lead contamination problem, with an estimated 90th percentile of 27 μg/L (first draw) for a back-calculated legitimate USEPA LCR monitoring event with 50% lead pipes (Pieper et al., 2018). If it is assumed that the 2.8X higher lead in biosolids in June 2014 vis-à-vis August 2015 is directly proportional to overall 90th percentile lead in water at that time [as supported by the strong correlation in other time periods; Biosolids-Pb = 0.37 x MWLL90 + 1.41 ($R^2 = 0.66$; $p < 0.05$; one-sided t-test for Pearson r)], it would suggest a 90th percentile lead first draw in the range of 76.8–98.5 μg/L in the worst (outlier) month of the FWC (June 2014) versus the EPA action level of 15 μg/L.

But first draw lead is only one part of the human exposure picture. The 2001–2004 DC Lead crisis was noteworthy because second draw was often much higher than first draw (Edwards and Dudi, 2004; Edwards et al., 2009; Edwards, 2013), whereas in Flint, the opposite was true (Pieper et al., 2018). If a simple composite exposure is considered based on 50% first draw and 50% second draw weighting (MWLL90), the 62.4 μg/L in the outlier month of June 2014 of the FWC would be in the range of the water lead observed in the 2001–2004 DC Lead Crisis (Fig. 4), but the DC exposures were much worse at all other times and the exposure was

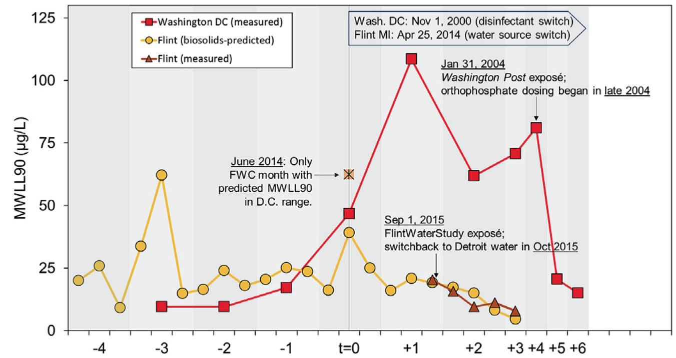

Fig. 4. Calculated MWLL90 levels from Washington D.C. lead in drinking water crisis, compared to estimated MWLL90 levels during the Flint, MI water crisis (FWC) as predicted from biosolids lead (this paper; Fig. 2) or that measured during five citizen science sampling rounds (method of Pieper et al., 2018). Washington D.C. data is yearly for 1997–2006 with t = 0 being the year 2000, while predictions based on Flint biosolids data is averaged over 4-month intervals (Dec–Mar, Apr–Jul, Aug–Nov) for Dec 2009–Jul 2015 and t = 0 represented by Apr-Jul 2014. Citizen science FWC sampling months were Aug 2015, Mar 2016, Jul 2016, Nov 2016, Jul 2017 at t = +1 to +3 years). For FWC, June 2014, is the only month where biosolids-predicted MWLL90 was in the range of the D.C. crisis.

of much longer duration (Edwards et al., 2009; Edwards, 2013; Roy and Edwards, 2019a). Specifically, the MWLL90 for the FWC, without considering the outlier month of June 2014, ranged from 10.2 μg/L to 43.1 μg/L, whereas that for Washington D.C. 2001–04 were between 61.9 μg/L and 108.6 μg/L. This is expected, since the D.C. lead crisis elevated a significant percentage of children's blood lead above the 10 μg/dL CDC “level of concern” in force at that time, whereas the FWC was manifested at the %EBL5 level and not at > 10 μg/dL (Edwards et al., 2009; Edwards, 2013; Hanna-Attisha et al., 2016; Gómez et al., 2018). Future studies examining possible public health harm from lead and other metals released from the plumbing during the FWC, should also carefully consider results indicating June–August 2014 was the time period of maximum water lead exposure.

3.6. Innovative use of biosolids monitoring data as a cumulative measure of WLLs

Urban sewage is being increasingly monitored to identify and map general public health trends from antibiotic resistance genes (ARGs), pharmaceutical and personal care products (PPCPs) and population-level traits like obesity and the human gut microbiome (Cai et al., 2014; Newton et al., 2015; Olofsson et al., 2012; Su et al., 2017; Wang and Wang, 2016). This study suggests that biosolids monitoring can provide important insights about overall trends in lead release to water from plumbing, which is important given rising worldwide concern about exposure to lead in drinking water, and the logistical and statistical problems of monitoring lead at consumer taps (Roy and Edwards, 2019a).

Targeted sampling, random sampling, 3-D profiles and proportional sampling of drinking water in consumer homes – all have acknowledged strengths and weaknesses, proponents and detractors (Clark et al., 2014; Del Toral et al., 2013; Masters et al., 2016a; Pieper et al., 2017; Schock, 1990; Jarvis et al., 2018; Lytle et al., 2019; Riblet et al., 2019), and, in some cases, a hundred household samples are required by regulation to calculate 90th percentile lead and monitor effectiveness of corrosion control for just one month each year. If a single sample of sludge could be used to track aggregate lead release and corrosion control effectiveness every month, as seems to have been the case in this research, it could improve understanding of seasonal trends and problems

with semi-random particulate lead release plaguing at the tap analysis of LCR monitoring data.

Specifically, biosolids lead monitoring may provide highly complementary, if not some superior, insights to traditional approaches that rely on direct monitoring of lead in drinking water at consumer taps. This is an exciting prospect deserving of future study that even seems obvious in retrospect given prior understanding. It is especially important considering the cost, logistical problems of accessing sampling taps in home and buildings for compliance sample collection, and the hundreds (or even thousands) of samples that would be required to obtain statistically valid estimates of water lead regulatory goals (i.e., 90th percentile lead in the U.S.) as indicated by prior research (Masters et al., 2016a).

4. Conclusions

Our novel approach shedding light on WLLs during the FWC based on routine biosolids analysis revealed that:

- Plumbing-related metals, including lead, were strongly correlated with one another in monthly sewage biosolids monitoring data during 2011–17, especially during the FWC months of Apr 2014–Oct 2015.
- The plumbing related metals Cu, Zn and Pb were also correlated with one another in calculated weighted averages of first, second and third draw, in five rounds of Virginia Tech and USEPA drinking water monitoring data.
- Biosolids lead strongly correlated with citywide WLLs in Virginia Tech's sampling from August 2015 to August 2017.
- During the FWC, the increased biosolids lead mass (≈ 23 kg) was just 14% higher than the comparable 18-month time period pre-FWC, but most (76%) of that increased mass was in the months of July–September 2014. During those three months %EBL5 was nearly doubled ($p < 0.05$) during FWC versus pre-FWC, but was not significantly higher in the other months of the FWC.
- Biosolids lead was very weakly correlated with %EBL5 pre-FWC and during the FWC, and not at other time periods, consistent with the expectation that water lead exposure is not strongly correlated to blood lead.
- Lead filters and bottled water severed the link between biosolids lead and %EBL5, consistent with public health protections of Flint consumers during the Federal Emergency.
- Exposure to elevated water lead during the FWC was predominantly associated with a large lead release that occurred during summer 2014, as evidenced by high lead in biosolids and %EBL5 in children. This is consistent with prior research based only on %EBL5.
- Summer spikes of WLL occurred when orthophosphate was not added to water in 2014 and 2015, but not in pre-FWC or post-FWC summer months when orthophosphate was being dosed.
- Higher orthophosphate dosages resulted in lower WLLs and biosolids lead levels, demonstrating the effectiveness of increased phosphate dosing.
- Biosolids lead monitoring may provide unique insights to effectiveness of lead corrosion control and citywide exposure risks.
- Biosolids lead and predicted human water lead exposures, during the 2014–2015 FWC, were in the range of what occurred in 2011.

Declaration of interests

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

> Aside from our work exposing the Flint water crisis, our data and testimony have been subpoenaed in several Flint water-related lawsuits. We are not party to any of the lawsuits. Dr. Edwards has been subpoenaed as a fact witness in many of the lawsuits, but he has refused all financial compensation for time spent on those activities. Previously, Dr. Edwards served as an uncompensated fact witness in lawsuits pertaining to Washington DC lead-in drinking water crisis, but these lawsuits have ended.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2019.05.091.

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